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Magnetic microstructure of the Finemet-type thin films

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Abstract. Fe_{73.9}Cu₁Nb₃Si_{13.2}B_{8.9} thin films were deposited on the glass substrates by means of ion-plasma radio frequency sputtering. Heat treatment at temperatures of 350, 400 and 450 °C was performed for 30 minutes in order to control thin film structural state. The X-ray powder diffraction showed that the crystallization of α -Fe(Si) nanograins took place at the temperature of 420 °C whilst the other samples stayed in the amorphous state. The magnetic microstructure of the Fe_{73.9}Cu₁Nb₃Si_{13.2}B_{8.9} films was discussed in the framework of random anisotropy model and law of approach of magnetization to saturation. The parameters of magnetic microstructure were determined, and relations between structure and macroscopic magnetic properties were interpreted by means of magnetic microstructure analysis.

1. Introduction

Nanocrystalline Fe_{73.9}Cu₁Nb₃Si_{13.2}B_{8.9} (Finemet) alloy is well known as soft magnetic material with low coercivity and both high saturation magnetization and permeability [1]. Macroscopic properties of Finemet alloy are closely related to the microscopic structural and magnetic parameters. This relationship has been successfully described within the framework of random magnetic anisotropy model (RAM) [2].

It is believed that the structure of amorphous and nanocrystalline materials determines the formation of a magnetic microstructure, which in turn determines their magnetic properties. In such materials, according to the random anisotropy model, the magnetic microstructure is a set of stochastic magnetic domains, in each of which the ferromagnetic order is realized due to the strong exchange interaction. Within this model, the characteristics of a stochastic domain, such as its size and anisotropy, is determined by local structure parameters (exchange parameter, local anisotropy constant, and size of structural element (particles, grain, cluster, etc.)).

In particular, it was found that the averaged magnetic anisotropy constant $\langle K \rangle$, which characterizes the stochastic magnetic domain, determines the value of the coercivity H_C and the magnetic permeability μ [3]. The $\langle K \rangle$ value is the indicator of local magnetic anisotropy (K) averaging efficiency inside the stochastic magnetic domain. Thus, the study of magnetic microstructure can provide useful information about the internal microstructure of the material, the processes of magnetization and magnetization reversal, and, in the case of thin films, the degree of influence of the ferromagnetic correlations' dimensionality on the macroscopic properties.

2. Samples and experiment

2.1. Samples preparation



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Thin films with a thickness of 200 nm were synthesized by means of a high-frequency ion-plasma sputtering technique. Sputtering conditions were characterized by the presence of the 99.987 % argon atmosphere and the permanent magnetic field of 100 Oe that was applied in the film plane, and the pressure of residual gases in the sputtering chamber and the argon working pressure were about 10^{-6} and 10^{-3} Torr correspondingly.

The nominal composition of the films was $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$. The films were deposited on the borosilicate glass (Wilma LabGlass©) substrate. The substrate temperature during the deposition was 40–50 °C.

The films' structure was modified using a heat treatment implemented in the sputtering chamber. The heat treatments were performed at the temperatures of 350, 400 and 450 °C during 30 minutes. Heating was done using an infrared lamp. The temperature was measured near the film surface using a thermocouple. The range of fluctuations of an established annealing temperature did not exceed ± 5 °C. The heat treatment conditions regarding the pressure and magnetic field were the same as during the deposition.

2.2. Experimental techniques

The thickness of the films was verified by the height difference between the film and the substrate using Dektak 150 Stylus Profilometer. The structure of the as-prepared sample and of the samples after annealing at 350, 400 and 450 °C was studied by means of the Bruker D8 Discover diffractometer with the $\text{Cu-K}\alpha$ radiation (radiation wavelength was $\lambda=1.5418$ Å) and a graphite monochromator. The measurements were taken in the 2θ angle range from 10 to 125° at the room temperature. The XRD patterns were processed using the TOPAS 3 program with the Rietveld algorithm for the refinement of structural parameters, and the crystallite size was determined from the XRD patterns using the Selyakov–Scherrer formula [4].

Temperature dependences of magnetization ($M(T)$) and magnetization curves ($M(H)$) were measured using a magnetic properties measurement system MPMS XL7. The applied magnetic field during the $M(T)$ measurements was 100 Oe. $M(H)$ dependencies of the films were taken at the room temperature in the field of up to 50 kOe.

The hysteresis properties of the films underwent various heat treatments were investigated by means of a magneto-optical microscope Evico Magnetics GmbH. The external magnetic field was applied along the easy magnetization axis (EA) of the samples.

The atomic force (AFM) and magnetic force (MFM) microscopy images were obtained using the Scanning Probe Microscope Bruker Multimode 8. The MFM mode of measurements is based on a two-pass scanning technique. The sample topology was recording in the tapping mode for the first pass. In the course of the second pass, magnetic phase contrast was measured in the lift mode (typical height about 80-100 nm). The scans were performed in the presence of an external magnetic field ($H \sim 180$ Oe) directed out-of-plane to the sample surface. The scan area was $10 \times 10 \mu\text{m}^2$ (resolution – 512×512 pixels) and the scan rate was 0.150 Hz. Tip with an approximate radius of 40 nm, magnetic CoCr coating, and spring constant of 5 N m^{-1} was used in experiments.

2.3. Model

The data of magnetic measurements were processed within the framework of the approach developed by V.A. Ignatchenko, R.S. Iskhakov and S.V. Komogortsev [5-7] based on the analysis of the random anisotropy model (RAM) [3] and the law of approach of magnetization to saturation (LAMs) [8]. This approach allows estimating the magnetic microstructure parameters (local magnetic anisotropy field (H_a), magnetic anisotropy correlation radius (R_c), averaged magnetic anisotropy field ($\langle H_a \rangle_L$) and ferromagnetic correlation radius (R_L)) and the interrelationship between the nanocrystalline structure and magnetic properties.

The relationship between the structural state and the magnetic properties of amorphous and nanocrystalline magnetic materials was demonstrated using the random anisotropy model. In this model, the magnetic microstructure in disordered magnets is originated because of the competition

between fluctuating local magnetic anisotropy and exchange interaction. As a result, the regions of ferromagnetic ordering are formed and characterized by a finite ferromagnetic correlation length (R_L). For an ensemble of such no interacting stochastic magnetic domains, the law of approach of magnetization to saturation is valid, that allows extracting the averaged characteristics of the magnetic and structural correlation regions. According to this law, above a certain critical field H_R , the field dependence in the region of approaching of magnetization to saturation obeys the Akulov law $M \sim H^2$, and below – the critical field $M \sim H^n$, where $n < 2$ and depends on the spatial dimension of the random anisotropy axes distribution. The law of approach of magnetization to saturation can be written as

$$\langle M_z \rangle / M_S \approx 1 - d_m(H) \quad (1)$$

Here $d_m(H)$ is dispersion of the transverse component of magnetization. In equation (1) $\langle M_z \rangle$ and M_S are average magnetic moment in z -direction and saturation magnetization correspondingly. The following relations determine the form of the field dependence of the dispersion of magnetization in the approach of magnetization to saturation region

$$d_m(H) = (aH_a)^2 \cdot \begin{cases} H_R^{-d/2} H^{-(4-d)/2}, & H \leq H_R \\ H^{-2}, & H \geq H_R \end{cases} \quad (2)$$

In equation (2) a is symmetry coefficient [7], H_R is correlation field corresponding to the change of $d_m(H)$ type and containing information on R_L and R_C , and d is dimension of the magnetization correlations.

3. Results and discussion

The structure and the phase composition of the films were studied by means of XRD. In Figure 1, the diffraction patterns of the 200 nm films in the as-prepared state and after 30 min annealing are presented. The structure of the as-deposited film can be characterized as amorphous. The crystallization starts at the temperature of about 420 °C. The crystalline phase consists of the α -Fe(Si) grains with a size of about 16 nm. The grains size does not change as annealing temperature raises, but the volume of the crystalline phase increases. After the heat treatments at temperatures of 420 and 450 °C, the crystalline phase coexists with the amorphous matrix, thus the nanocrystalline state is realized. The heat treatment at 500 °C leads to fully crystallized film.

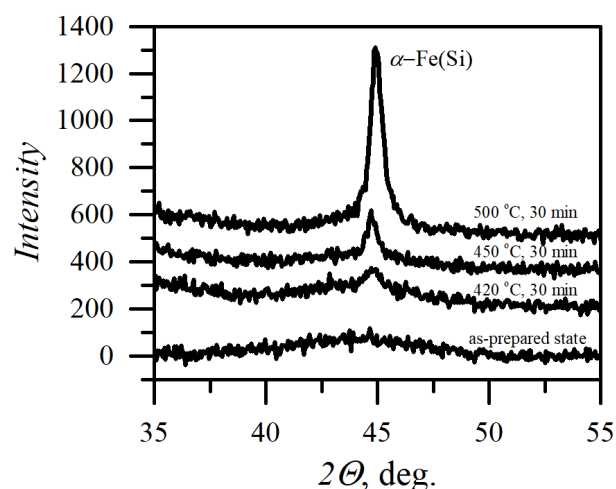


Figure 1. XRD patterns of 200 nm films of $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ in the as-prepared state and after annealing at 420, 450 and 500 °C for 30 minutes.

The magneto-optical hysteresis loops of the $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ films in the as-prepared state and after the heat treatment at 350, 400 and 450 °C are shown in Figure 2. Taking into account films XRD data it is seen that the hysteresis loop shape and the coercivity of the films undergo changes as a result of the heat treatment. The coercivity of the films slightly increases and the shape of the hysteresis loops becomes less rectangular after the annealing at 350 °C. With a subsequent increase in temperature up to 400 °C, a decrease in the coercivity is observed. After the heat treatment at 450 °C, the coercivity of the film increases dramatically.

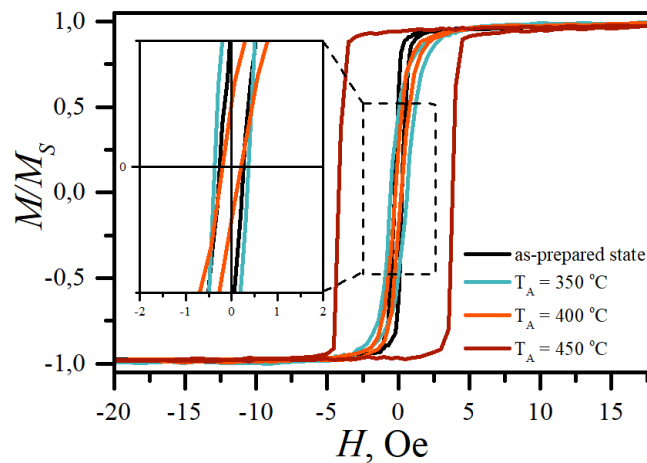


Figure 2. Magneto-optical hysteresis loops of 200 nm $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ films in the as-prepared state and after annealing at 350, 400 and 450 °C for 30 minutes.

The amorphous structure of the as-prepared films is characterized by large internal stresses. As can be seen from the hysteresis loops (Figure 2), after the heat treatment at 350 °C a change in hysteresis loop form takes place, which indicates a relaxation of internal stresses. After annealing at 400 °C the coercivity value decreases and is closed to the coercivity of as-prepared film. Based on the XRD study results, it can be assumed that at the temperature of about 400 °C the film is at the stage of the onset of crystallization, and their structure can be characterized by phase fluctuations. This structural instability can lead to variations in the coercivity value depending on the film sample. The noticeable differences in the hysteresis loops of the films are made oneself conspicuous after annealing at 450 °C. The coercivity increases. Because of crystallization, there is a difficulty in the magnetization reversal process and a decrease in the residual magnetization, that is, the loop loses its squareness. Thus, there is a relationship between the data of XRD analysis and hysteresis properties.

According to the Random anisotropy model the magnetization process is closely related to the average anisotropy $\langle K_L \rangle$, averaged over the region R_L . There are relationships that allow us to establish the averaged characteristics of the magnetic microstructure. Knowing the values of the exchange parameter A , anisotropy constant K and magnetic anisotropy correlation radius R_C , one can analyze the influence of the structural state of the thin films on their magnetic microstructure within the framework of the random anisotropy model. R_L and $\langle K_L \rangle$ are determined by the expressions

$$\langle K_L \rangle = K \left(\frac{R_C}{R_L} \right)^{\frac{2d}{4-d}}, \quad (3)$$

$$R_L = \frac{(R_c^*)^{4/4-d}}{R_c^{d/4-d}}, \quad (4)$$

where $R_c^* = \sqrt{A/\varepsilon K}$ is the critical radius of the homogeneous orientation of the anisotropy axes ($\varepsilon = 0.4-0.7$ is the numerical coefficient) [7].

The exchange parameter can be determined experimentally from the temperature dependence of the magnetization using the Bloch law ($T^{3/2}$). By means of LAMS, anisotropy constant K can be estimated. For this purpose, the temperature dependences of the magnetization and the curves of the approach of magnetization to saturation of the films with a thickness of 200 nm were obtained (Figures 3(a, b)). Exchange parameters of the as-prepared film and films annealed at 350, 400 and 450 °C determined from the temperature dependence of the magnetization were about $4.8 \cdot 10^{-7}$, $4.9 \cdot 10^{-7}$, $5.3 \cdot 10^{-7}$, $6.6 \cdot 10^{-7}$ erg cm⁻¹, correspondingly. The magnetization curves were rearranged in double logarithmic coordinates in the form of the dependence of the dispersion of magnetization d_m on the magnetic field H . This type of curves allows to uniquely determine the parts of curves corresponding to the theoretical power dependences of the dispersion of magnetization on the magnetic field (Figures 3(c, d)).

In Figures 3(c) the $d_m(H)$ dependences have two areas that can be identified for analysis - relatively low and high fields. When considering the low-field part of the curves (up to 200 Oe), it can be seen that the magnetization process significantly depends on the heat treatment conditions. That is, in this field area, even small changes in the films' microstructure affect the magnetization process. However, in this case interest is in high-field (above 30 kOe) part of the curve. The similar character of the curves (in the field above 1 kOe) of films in the initial and annealed at 350 °C states indicates the presence of similar local magnetic microstructural parameters. Structural changes in the film annealed at 400 °C are associated with almost complete relaxation of internal stresses and the beginning of the formation of precursors of the crystalline phase. With such a relaxed structural state, a rapid decrease in the dispersion of magnetization can be expected in the high-field region of the curve, which is observed experimentally. After the heat treatment at 450 °C, a difficulty of a decrease in the dispersion of magnetization is observed, which indicates an increase in local magnetic anisotropy.

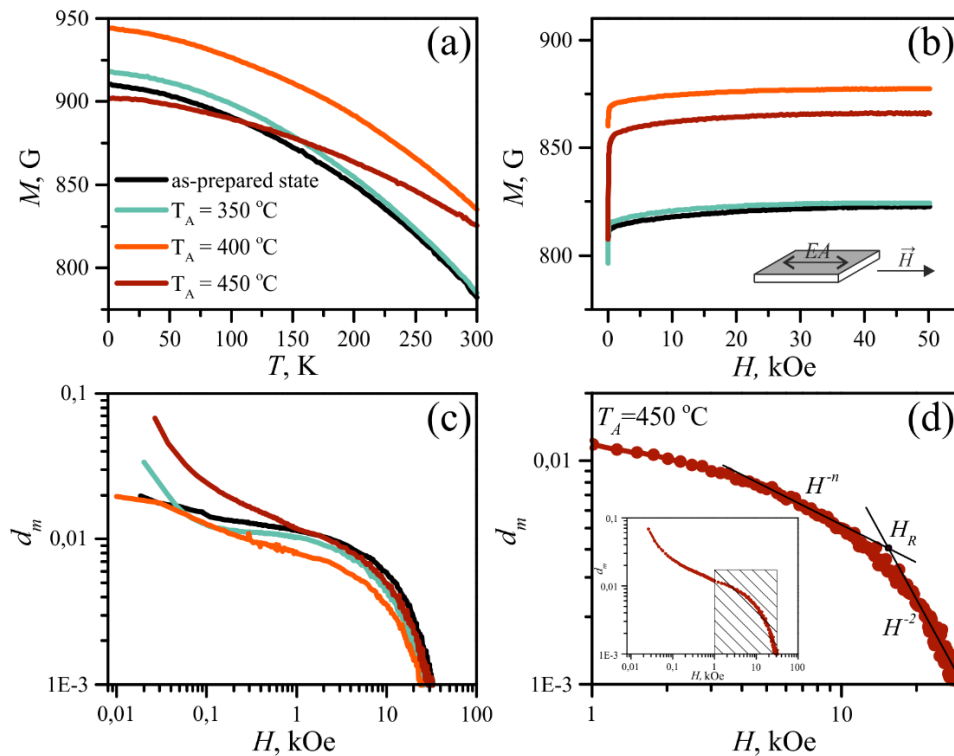


Figure 3. $M(T)$ (a) and $M(H)$ (b) dependencies, d_m dependence in log-log coordinates (c), an example of search of the curve parts corresponding to the power regimes (d).

The presented dependences allow one to quantify the characteristics of structural and magnetic correlations and to analyze in detail the processes occurring in the thin films in the process of preparation and heat treatment under various conditions.

On each of the dependences $d_m(H)$, constructed in double logarithmic coordinates (Figures 3(c)), a search for linear dependencies corresponding to the power regimes indicated above in equation (2) was performed.

Figures 3(d) shows the high-field part of the dispersion magnetization dependence of a film after annealing at 450 °C. In this figure, asymptotes are shown by straight lines, corresponds to the power dependency of the magnetization dispersion as required by equation (2). Their intersection determines the magnitude of the field H_R . Approximation of linear part on the dispersion of magnetization curves, for which the $d_m \sim H^2$ (under the condition $H \geq H_R$), allows estimating the local anisotropy field H_a . As a result, the local anisotropy constant K was determined, and its value was about $1\text{--}2 \cdot 10^6$ erg cm⁻³ depending on the structural state of the films. The smallest value was for the film after the heat treatment at 400 °C (Table 1).

With annealing temperature increasing up to 400 °C, the structural changes are accompanied by an increase in the correlation radius of the local anisotropy axes, saturation magnetization, exchange parameter, magnetic correlation radius, at the same time the local and average magnetic anisotropy constants decrease. In this case, the most drastic change in the parameters of the structure and magnetic microstructure of the film occurs after annealing at 400 °C. According to the study of hysteresis properties (Figure 2), this film has the best magnetic properties, demonstrating the smallest value of the coercivity. The structural state of this film is not strictly defined. However, on the basis of a comprehensive study, it can be assumed that it is characterized by complete stress relaxation and the onset of the crystallization process.

Table 1. Parameters of the magnetic microstructure of $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ films with thickness of 200 nm.

State of specimen	R_C (nm)	K (10^6 erg cm^{-3})	R_L (nm)	$\{K_L\}$ (10^6 erg cm^{-3})
As-prepared state	2.7	1.7	160	3.6
350 °C	2.8	1.5	200	2.5
400 °C	2.9	1.1	365	0.8
450 °C	3.1	1.8	190	3.8

With annealing temperature increasing up to 400 °C, the structural changes are accompanied by an increase in the correlation radius of the local anisotropy axes, saturation magnetization, exchange parameter, magnetic correlation radius, at the same time the local and average magnetic anisotropy constants decrease. In this case, the most drastic change in the parameters of the structure and magnetic microstructure of the film occurs after annealing at 400 °C. According to the study of hysteresis properties (Figure 2), this film has the best magnetic properties, demonstrating the smallest value of the coercivity. The structural state of this film is not strictly defined. However, on the basis of a comprehensive study, it can be assumed that it is characterized by complete stress relaxation and the onset of the crystallization process.

After annealing at 450 °C, crystallization occurs in the films. This leads to a noticeable increase in the exchange parameter and the local anisotropy constant, and the saturation magnetization decreases. Increased during crystallization, the magnetic anisotropy of α -FeSi grains, together with their random orientation, leads to a sharp increase in the magnetization dispersion, which is clearly seen in Figure 3(c) in the fields below 1 kOe.

From the magnetic properties analysis and data in Table 1, it follows that the greatest value of ferromagnetic correlation length (R_L) corresponds to the smallest value of coercivity. It means that the larger the area inside which the averaging of the local anisotropy axes occurs, the easier the process of magnetization reversal is.

The experimentally magnetic microstructure can be found in the form of inhomogeneities of magnetic contrast in images obtained using a magnetic force microscope.

Figure 4(a, b) shows the surface topography and the image of the magnetic contrast of 200 nm film after annealing at 450 °C. Regions of homogeneous contrast on MFM images can be correlated with the length of the magnetization correlation region. The image as a whole can represent a stochastic magnetic domain structure. The heterogeneity of the magnetization and color contrast on the MFM image are not uniquely related, but the statistical properties and the spatial scales characteristic of the inhomogeneities must be the same.

It is seen in Figure 4(c) that the correlation length, calculated from MFM images, is anisotropic in nature. This is typical for polycrystalline films [9]. It should also be noted that the size of the regions of correlation is much larger than the thickness of the films. This will impose some additional anisotropy on their shape along the direction perpendicular to the film plane. The correlation radii of magnetization of R_L^{ox} and R_L^{oy} films, defined as the size at which the correlations halve, correspond to the shortest and longest correlations of magnetic contrast in two mutually perpendicular directions ox

and oy . The mean correlation length was estimated as $R_L = (R_L^{ox} \cdot R_L^{oy})^{-\frac{1}{2}}$. The value of the correlation length calculated in this way is about 400 nm, which is in order of magnitude consistent with the correlation length determined using LAMS and RAM.

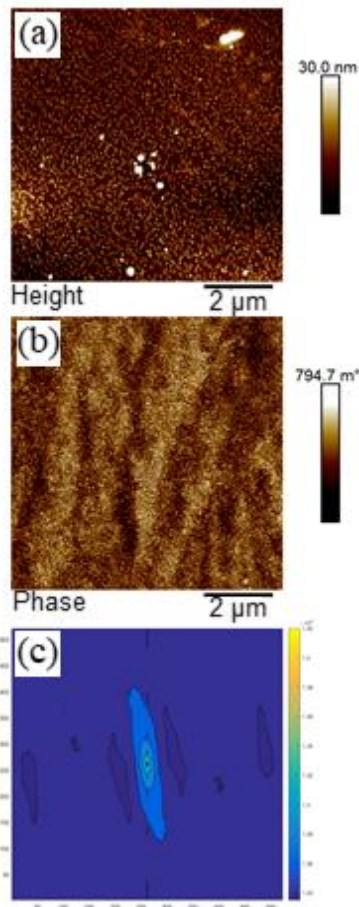


Figure 4. AFM (a) and MFM (b) images and correlation length images (c), culculated from MSM images, of 200 nm film.

4. Conclusion

In that work magnetic microstructure of the $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ thin films was investigated. Magnetic microstructure parameters were determined using the random anisotropy model, the law of approach of magnetization to saturation, and analysis of inhomogeneities of magnetic contrast on magnetic force microscopy images. These approaches allow getting a fairly complete picture of the relationship "structure – magnetic properties" of the studied films.

The best soft magnetic properties of the $\text{Fe}_{73.5}\text{Nb}_3\text{Cu}_1\text{Si}_{13.5}\text{B}_9$ thin films are realized after the heat treatment at 400 °C. The structural state of this film is predominantly amorphous but is characterized by phase fluctuations. Based on the magnetic properties, it can be assumed that the structure is fully stress relaxed. The averaging of the local magnetic anisotropy axes by the exchange interaction is the most efficient for the film annealed at 400 °C. Hence, $\langle K_L \rangle$ has minimal value. In accordance with $\langle K_L \rangle$ is directly proportional to H_C , it is expected that coercivity is also minimal, what was observed in the experiment.

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